Applied Industrial Hygiene

ISSN: 0882-8032 (Print) (Online) Journal homepage: https://www.tandfonline.com/loi/uaph20

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To cite this article: Bruce Wilbur Hills & A. L. Greife (1986) Evaluation of Occupational Acrylamide Exposures, Applied Industrial Hygiene, 1:3, 148-152, DOI: 10.1080/08828032.1986.10390499

To link to this article: https://doi.org/10.1080/08828032.1986.10390499

Evaluation of occupational acrylamide exposures

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Since commercial production of acrylamide began in 1954, there have been at least 60 reported cases of acrylamide poisoning resulting in peripheral neuropathy in workers. All of these cases except one have involved occupational exposure to acrylamide monomer from either acrylamide and/or polyacrylamide manufacturing, or acrylamide grouting. Because of a lack of adequate exposure data for these reported cases of acrylamide intoxication and for current work environment conditions, investigators from the National Institute for Occupational Safety and Health have undertaken a study to evaluate exposures in four acrylamide-polyacrylamide manufacturing plants and at one sewer grouting site. Ninety-one personal eight-hour time-weighted average (TWA) exposures from 81 workers in the manufacturing plants had airborne acrylamide levels ranging from 0.001 to 0.392 mg/m^3 with a geometric mean of 0.036 mg/m^3. The mean exposure level for monomer operators was 0.065 mg/m^3; polymer operators, 0.031 mg/m^3; and maintenance personnel, 0.013 mg/m^3. A greater range of exposures occurred between the manufacturing plants. One facility, which produced a dry and an aqueous acrylamide, had a geometric mean acrylamide exposure level 16 times higher than the level found at another slightly smaller plant that produced acrylamide in solution. At the sewer-line repair site, where two workers applied an acrylamide grout, acrylamide exposures were only 0.002 and 0.007 mg/m^3. Although the sewer repair workers had very low inhalation exposures, they were at high risk of being dermally exposed to acrylamide. **Hills, B. W.; Greife, A. L.**: Evaluation of occupational acrylamide exposures. *Appl. Ind. Hyg. 3*:148-152; 1986.

Introduction

Acrylamide monomer is a white, crystalline solid which is highly soluble in water and has a very low vapor pressure. It is a reactive monomer that contains an amide group and a double bond vinyl group, which allows it to readily undergo polymerization and copolymerization reactions.[1]

An estimated 90% of all domestically produced acrylamide monomer is used for the production of polyacrylamides. A number of different types of polymers can be formed by modifying the polymerization reaction to create the desired properties. These polymers have applications including separation and clarification of liquid-solid phases and also are used for binding, thickening, lubricating, and film formation. These properties are beneficial in the treatment of waste water and drinking water and are used in the pulp and paper, mining, construction, petroleum, textile, cosmetic, and food industries.[1,2] The remainder of domestically produced acrylamide monomer is further reacted to form N-substituted acrylamides such as N-methylolacrylamide, and a portion is used as a chemical intermediate in the coating-finishing and adhesives industries.

Another major use of acrylamide is in the application of acrylamide grout for sewer line repair and soil stabilization.[1,3] This process involves injecting the monomer into the ground with the catalyst, and polymerization occurs forming a gel-like, water impermeable seal. At least 90% of all acrylamide grouting is used to repair sewer pipes to prevent the infiltration of ground water. The remaining 10% of grouting is performed by the construction and mining industry to stabilize soil to improve formation strength. At present, all acrylamide used in this country for grouting is imported as a dry product.

Toxicity and case studies

Monomeric acrylamide is a neurotoxin which affects the peripheral and central nervous system. It also causes skin damage, such as erythema and peeling of exposed areas of the skin after dermal contact with acrylamide.[4] Neurological symptoms first appear approximately one to two months after a toxic exposure. Chronic exposures to relatively low amounts of the monomer will induce symptoms, but there is little occupational exposure data to determine at what dose this occurs in humans. Early symptoms are sweating, erythema and peeling skin on the hands and feet, muscle weakness, sensory loss, coldness, numbness of the lower limbs, burning sensation, tenderness to the touch, and the absence of reflexes.[4] Later symptoms include loss of body weight, lassitude, sleepiness, emotional changes, loss of vibration and position senses, positive Romberg's sign, weak or absent tendon reflexes, muscular atrophy, foot drop, unsteady walk, and occasional urinary and fecal retention.[4] Recovery from these symptoms does occur but may take months to several years. Exposure to acrylamide can occur by absorption through the skin, through the eyes, by ingestion, or inhalation as a dust, vapor, or aerosol

0882-0832/86/030148-152$2.00

TABLE I
Case studies report

Year	No. of cases	Exposure	Location	References
1953-54	5-6	monomer mfg.	U.S.	(10)
1961	10	monomer mfg.	Japan	(11)
1967	1	grouting	Canada	(12)
1967	6	polymer mfg.	England	(13)
1967	1	polymer mfg.	Italy	(14)
1969	4	monomer mfg.	France	(15)
1970	1	grouting	France	(16)
1971	15	polymer mfg.	Japan	(17)
1971	3	unknown	Japan	(18)
1972	1	grouting	France	(19)
1975	5	well water	Japan	(9)
1976	1	polymer mfg.	U.S.	(20)
1977	6	grouting	England	(21)

solution. Polyacrylamide has not been shown to be toxic, but the polymer does contain unreacted monomer.[4,5] The percentage of monomer in the polymer is usually below 0.1%, but it has been as high as 5% in the early 1970's.[6-8]

Case reports of human toxicity all have been associated with industrial exposure except one incident in Japan where well water was contaminated. Five individuals developed mental confusion, muscle incoordination, and later the symptoms and signs of polyneuropathy after sewer lines were repaired with an acrylamide grout adjacent to wells used for drinking water and bathing.[9] All other cases (Table I) occurred in the manufacturing of acrylamide and polyacrylamide and during soil stabilization.[10-21] The major route of exposure appears to be dermal absorption of the solution and inhalation of dry monomer.

In 1976, the National Institute for Occupational Safety and Health recommended that workers not be exposed to airborne acrylamide levels above 0.3 mg/m^3 as a 10-hour time-weighted average.[22] This recommended level (like the permissible exposure limit of the Occupational Safety and Health Administration and the Threshold Limit Value of the American Conference of Governmental Industrial Hygienists of 0.3 mg/m^3 as an 8-hour time-weighted average) was developed to protect workers from acrylamide's neurotoxic, eye, and skin damaging properties.[23,24] Because of the reported carcinogenicity of acrylamide in laboratory animals, the American Conference of Governmental Industrial Hygienists intends to change its acrylamide Threshold Limit Value to 0.03 mg/m^3 as an 8-hour time-weighted average.[25]

Since 1976, new information on the carcinogenic potential of acrylamide has been reported. The manufacturers of acrylamide sponsored a two-year bioassay, in which groups of 90 male and female rats were given doses ranging from 0.01 to 2.0 mg acrylamide/kg/day in drinking water 5 days/week for 24 months. A statistically significant increase in numbers of tumors were observed in both sexes. In female rats, there were significant increases in tumors of the central nervous system, mammary gland, clitoral gland, uterus, oral cavity, pituitary gland, and thyroid gland. Significant increases in tumors were observed in the thyroid gland, adrenal gland, and scrotal cavity of male rats.[26] In another study, acrylamide was found to induce skin tumors in female mice and also found to induce lung adenomas in male and female mice using both the oral and intraperitoneal routes of administration.[27]

The authors of this study estimate that between 700 and 1000 workers are potentially exposed to acrylamide in the manufacturing of acrylamide monomer and polymer based on reporting from the acrylamide manufacturers. The NIOSH National Occupational Hazard Survey lists the estimated total number of U.S. workers who are potentially exposed to acrylamide to be approximately 10,000, employed in 27 occupations.[28] Workers performing acrylamide grouting are estimated to range from 600 to 1100, as based on the number of individuals who are licensed or certified by the grouting suppliers to perform acrylamide grouting.[29,30]

Description of workplace and methods

In this study, industrial hygiene surveys were conducted at all four domestic acrylamide manufacturing plants. Each of these plants also manufactures polyacrylamides. One survey was also conducted at a sewer line grouting site. The purpose of these surveys was to assess the extent and degree of worker exposure to acrylamide. Observations were made of work practices and the potential for dermal contact to dry and aqueous acrylamide. Area and personal air monitoring was performed to assess worker exposures to airborne acrylamide.

The starting material in the production of acrylamide is an aqueous solution of acrylonitrile which is hydrolyzed over a copper alloy catalyst. Unreacted acrylonitrile is recycled back into the reaction, and a 50% aqueous acrylamide solution is removed. From this solution, a dry acrylamide was being produced at one facility up until January of 1985. At present, all dry acrylamide is imported from Japan and England. At all four locations, a 50% acrylamide solution is produced that can be marketed or can be further reacted to form polyacrylamides and N-substituted acrylamides. The workers involved in the production of acrylamide and polyacrylamides can be grouped into the general job titles of monomer operators, polymer operators, monomer material handlers, polymer material handlers, maintenance workers, and utility operators. The specific job titles and duties vary, but these six job classifications cover the workers in the acrylamide monomer and polymer production areas. The monomer and polymer operators control the manufacturing process. A portion of their time is spent in the control room, and the remainder is spent in the process area regulating valves, checking gages, and collecting quality control samples. The material handlers load bags, drums, trucks, and rail cars with the final product. This can be in a solid or liquid form. Other workers in the acrylamide areas are the maintenance workers who repair or overhaul the production equipment and the utility operators who supply water, electric power, and steam to the plant.

During the survey at a sewer line repair site, two employees performed the repairs with the use of two service trucks. One service truck held the grouting equipment—hoses, pumps, mixing tanks, and video monitoring equipment—while the other truck contained hoses and a water supply. The

workmen first assembled the packer which was lowered into the manhole and then passed through the sewer. It was positioned by the use of cables and a video camera viewed from the truck's control panel. At the site of a leak, both ends of the packer were inflated, isolating the leaking joint. Above ground in the service truck, an employee poured the grouting material which is 95% acrylamide and 5% methylene-bis-acrylamide into a mixing tank containing water. The acrylamide was bagged with an inner liner bag that could be placed below the water level of the mixing tank. This prevented dust particles from escaping into the air. Acrylamide solution along with a catalyst was injected under pressure from the service truck via hoses to the center of the packer and forced into the surrounding soil. The monomer polymerized to form a water impermeable gel sealing the leak. Once the leak was sealed, the packer was deflated and moved to the next joint.

The sampling strategy for the study was designed to monitor all workers who were potentially exposed to acrylamide by collecting a single, full period eight-hour sample. The data were calculated to give individual values as an eight-hour time-weighted average (TWA) in order to compare exposures between manufacturing plants. In addition, two short term exposure samples (18 minutes) were collected at the sewer-line repair site from the individual who poured and mixed the dry acrylamide grout into the mixing tank. Wipe samples were collected as an indication of presence and possible sources of non-respiratory exposures.

The sampling and analysis was accomplished by collecting acrylamide vapor and particulate on a 37-mm mixed cellulose ester filter-pore size 0.8 μm, Millipore type AA, followed by a silica gel tube SKC No. 226-10, 100/50, at a flow rate of 1 liter per minute. The collected material was then desorbed from the collection media with water and analyzed by high performance liquid chromatography with an ultraviolet detector. The method was validated at air concentrations of 0.03 to 0.6 mg acrylamide per cubic meter of air based on a sample size of 480 liters of air. The limit of detection for the airborne and wipe samples was 0.6 μg acrylamide per sample.[31]

Wipe samples were collected as an indication of presence and possible sources of non-respiratory exposures. A mixed cellulose ester filter moistened with water was used to wipe a surface area of approximately 10 cm^2. The surfaces wiped included the exterior of gloves and hard hats, inside the face piece of respirators, desk tops in the control rooms, on cafeteria table tops, on laboratory bench tops, and on reactor vessels.

Results and discussion

The air monitoring data for the four manufacturing sites (plants A-D) and one grouting site (E) are summarized in Tables II, III, and IV.

Plant A had the greatest range of acrylamide air levels. This was due mainly to the production of a dry acrylamide. The highest levels were found in enclosed areas of the production building where workers would only occasionally enter with a full-face canister type respirator for acrylamide dust and vapors. Unlike plant A, plants B, C, and D have all or a portion of their production process equipment open to the ambient air. The acrylamide production equipment at plant B is entirely outdoors. The production equipment at plants C and D are within a building, but natural ventilation is provided by at least one open wall.

TABLE II
Summary of area air monitoring data for acrylamide

Location	No. samples	Range mg/m³	Geometric mean	GSD
A	19	0.014–8.291	0.269	3.146
B	18	0.003–0.157	0.029	2.193
C	18	0.004–0.009	0.006	1.180
D	28	0.001–0.015	0.010	2.482
E	2	0.001–0.009		

TABLE III
Summary of NIOSH personal exposure data for acrylamide

Location	No. samples	Range mg/m³ 8 hr. TWA	Geometric mean	GSD
A	27	0.001–0.392	0.080	2.210
B	18	0.015–0.132	0.046	1.694
C	19	0.003–0.012	0.005	1.470
D	26	0.001–0.113	0.013	2.685
E	2	0.002–0.007*		

*Nine-hour TWA

The monitoring results are also organized by job title in Table IV. Monomer operators at plants A, B, and D had a mean exposure level twice that of the polymer operators. Two utility operators at plant A had eight-hour TWA exposures above the OSHA standard. The company performed side-by-side sampling during the NIOSH survey and confirmed the utility operators exposure levels. The higher levels cannot be accounted for because their job duties do not involve direct exposure to acrylamide. Subsequent monitoring of these workers by the company revealed much lower exposure levels. Apparently further monitoring of utility operators is necessary to determine sources of exposure. Monomer material handlers can have brief, high exposures when loading trucks or rail cars with acrylamide solution if they fail to wear personal protective equipment. The workers observed in this study did wear full-face dust and vapor respirators, neoprene gloves, and an apron during this operation. During the study, only minor maintenance occurred at each facility. However, these maintenance workers may potentially be exposed to levels of acrylamide above the OSHA permissible exposure limit or exposed by dermal contact. At plant C, the major cleaning of the reactor equipment was performed by workers from a small contracting firm rather than in-house personnel.

Fifty wipe samples were collected from personal protective equipment and workplace surfaces where skin contact was most likely. Nearly all of the wipe samples were found to be non-detectable (below 0.6 μg acrylamide per sample), except for a wipe sample collected on the exterior of a polyacrylamide reactor vessel which had 30 μg/sample, a laboratory counter top which had 3.2 μg/sample, and a door handle which had 0.9 μg/sample.

At the sewer-line repair site, Location E, the two workmen performing the grouting were exposed to low levels of airborne acrylamide. Nine-hour per-

TABLE IV
Summary of NIOSH personal exposure data for acrylamide by job title

Job title	No. samples	Range mg/m³ 8 hr. TWA	Geometric mean	GSD
Monomer Operators*	19	0.001–0.227	0.065	2.051
Polymer Operators*	27	0.001–0.181	0.031	2.330
Monomer Material Handlers*	4	0.017–0.260	0.085	2.204
Polymer Material Handlers*	4	0.018–0.035	0.023	1.383
Maintenance	14	0.001–0.132	0.013	3.014
Utility Operators	4	0.004–0.392	0.116	2.501

*Does not include Plant C, because employees worked in both the monomer and polymer departments.

sonal air samples levels were 0.002 and 0.007 mg/m³. The two short term personal air samples, 18 minute duration, collected during the pouring and mixing of the grout did not detect any airborne acrylamide. This may be due to the high moisture content of the acrylamide and the method of pouring the grout. Area air samples collected inside the service truck and the company garage revealed 0.009 and 0.001 mg/m³ respectively.

Surface wipe samples collected from the control panel in the service van, on a safety cone placed on the road, and on the exterior of two rubber work gloves were free of acrylamide contamination. However, a wipe sample from the side of the grout mixing tank detected 44 μg/cm². The source of this contamination was apparent when a small amount of solid acrylamide was spilled on the tank and floor during one mixing. The spill was not cleaned up, and when the moisture in the grout evaporated, acrylamide became airborne as a dust or vapor.

Other poor handling procedures which can lead to increased dermal contact with acrylamide were observed; these included storing empty grouting bags and mixing cups on the floor, and only occasionally wearing rubber gloves when handling the packer or when washing the equipment. Another source of dermal acrylamide exposure is the inside of the work gloves. The gloves are intended to prevent skin contact with the grout. When distilled water was rinsed inside one of the gloves, the rinse water was found to contain 65 μg of acrylamide.

Conclusions and recommendations

In general, the results of this study have shown that the most important factor affecting a worker's exposure to airborne acrylamide is the facility or location where he works rather than his job duties. Examples of this can be seen from the geometric mean of area and personal air monitoring data between plants, Tables II and III. Plant C had personal mean exposures of 0.005 mg/m³, whereas the Plant A mean exposures were 0.080 mg/m³, or 16 times higher. Another general conclusion is that operators in the monomer areas have a mean exposure of 0.065 mg/m³ or twice as high as the operators in the polymer areas who have mean exposures of 0.031 mg/m³. There are situations where workers performing a certain task, such as loading acrylamide solution, bagging dry acrylamide, or repairing process equipment, can be exposed to elevated air levels of acrylamide. On the other hand, the study data show that workers at different plants with the same job title and similar duties were not consistently exposed at the same high levels. The sewer-line repair workers had very low exposures to airborne acrylamide because they worked outdoors.

In the acrylamide manufacturing plants, there appears to be a background or area acrylamide air level throughout the acrylamide areas of a plant. This background of airborne acrylamide varied widely in concentration between facilities. The reasons may be due to such factors as: 1. Plant design, 2. Production level, 3. Type of product (dry or wet), 4. Frequency of housecleaning, 5. Age and maintenance of equipment, 6. Use of engineering control equipment. 7. Use of natural dilution ventilation.

Wherever acrylamide is present in the plants, there is the potential for workers to be exposed via skin contact. This route of exposure can be controlled by the proper use of personal protective clothing such as rubber gloves, coveralls, non-leather soled work shoes, and rubber aprons. Inhalation exposure can be reduced by frequent washing of the acrylamide production areas with water and by supplying adequate workroom ventilation.

The acrylamide grouters appear to be at low risk of inhaling acrylamide but have a high risk of dermal exposure. Although there are no reported cases in the literature of toxicity to acrylamide in sewer line repair grouters, it is possible that mild symptoms of acrylamide toxicity may occur and go unreported. The latency period between a toxic acrylamide exposure and the first neurological symptoms is at least one month.[4] If any neurological symptoms of acrylamide toxicity were to occur, the workers might not relate their illness to their job. Unlike the manufacturing companies which have medical staff who are aware of acrylamide's toxicity and test employees for symptoms, the smaller grouting companies rely on local physicians who may not be as familiar with occupational diseases. The occupational health specialist who is responsible for assessing worker exposure to acrylamide during the grouting operation will need to understand each step of grouting application. Careful observations of each step in the handling of the grout is necessary to identify possible sources of dermal exposure.

Whereas most work sites examined in this study have acrylamide air levels below the current OSHA permissible exposure limit and NIOSH recommended levels, the dermal exposure hazard remains. An analysis of the studies of human intoxication with acrylamide indicate that dermal absorption and ingestion have been the main routes of exposure. In addition, current acrylamide evaluation criteria do not consider the recent information from animal studies on the carcinogenic effects of acrylamide. These factors should be taken into consideration by those with potential exposure to acrylamide.

Subsequent to the NIOSH surveys, the U.S. EPA and Midwest Research Institute (MRI) conducted four industrial hygiene surveys in the greater Washington, DC area of sewer line repair sites to further assess dermal exposures in these workers. Based on oral communication with the EPA-MRI survey team, and participation on two of the site visits by a NIOSH investigator, it was learned that interviews with 9 workers revealed that 5 of them described past symptoms of reddening

and skin peeling of the hands which is an early sign of acrylamide toxicity. One of the workmen observed had severe skin peeling of the hands. He reported past shortness of breath, muscular weakness of hands, arms, legs, feet, sweating, and fatigue. He also complained that his arms from the elbows down sometimes felt numb and that he had an aching in his legs and feet, but not as often as in his arms. He stated that his feet had given him problems even before he started grouting. His job as a grouter began five years ago and he has been employed by two different companies. A report on the results of this study will be available from the Office of Toxic Substances, U.S. EPA, TS-798 in the fall of this year.

Based on the surveys of acrylamide grouting operations, it is apparent that there is a large variation in the work practices of grouters. Some individuals follow safe handling procedures closely and wear personal protective equipment while others do not. Personal protective equipment appears to have an effect on the degree of dermal exposure, however, preliminary EPA-MRI results indicate that nearly all the grouters had hands contaminated with acrylamide despite the wearing of gloves. This was determined by conducting hand rinses on the workers and analyzing the rinse water. A possible explanation for the hand contact may be from periodic removal of the gloves and contact with contaminated equipment. The greatest determinant in dermal exposures is the type of grouting being performed. In the sealing of manholes, a worker must enter the manhole, drill holes in the leaking wall, and then pump acrylamide solution into the hole with an injection gun. Frequently the grouting solution will splash back at the workman resulting in his outer clothing becoming covered with the acrylamide. Rain gear is normally worn during this operation, yet the acrylamide solution was observed entering the inside sleeves of the rain jacket and wetting the arms. Evidently dermal contact with acrylamide in aqueous solution is still a problem in the sewer grouting work.

Acknowledgement

The authors wish to thank the U.S. Environmental Protection Agency for providing funding for this study under the NIOSH-EPA interagency agreement, number AD-75F-1555-0.

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Received 1/8/86; review/decision 2/12/86; revision 2/27/86; accepted 3/24/86